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An Examination of Some Micrometeorological Methods for Measuring Dry Deposition

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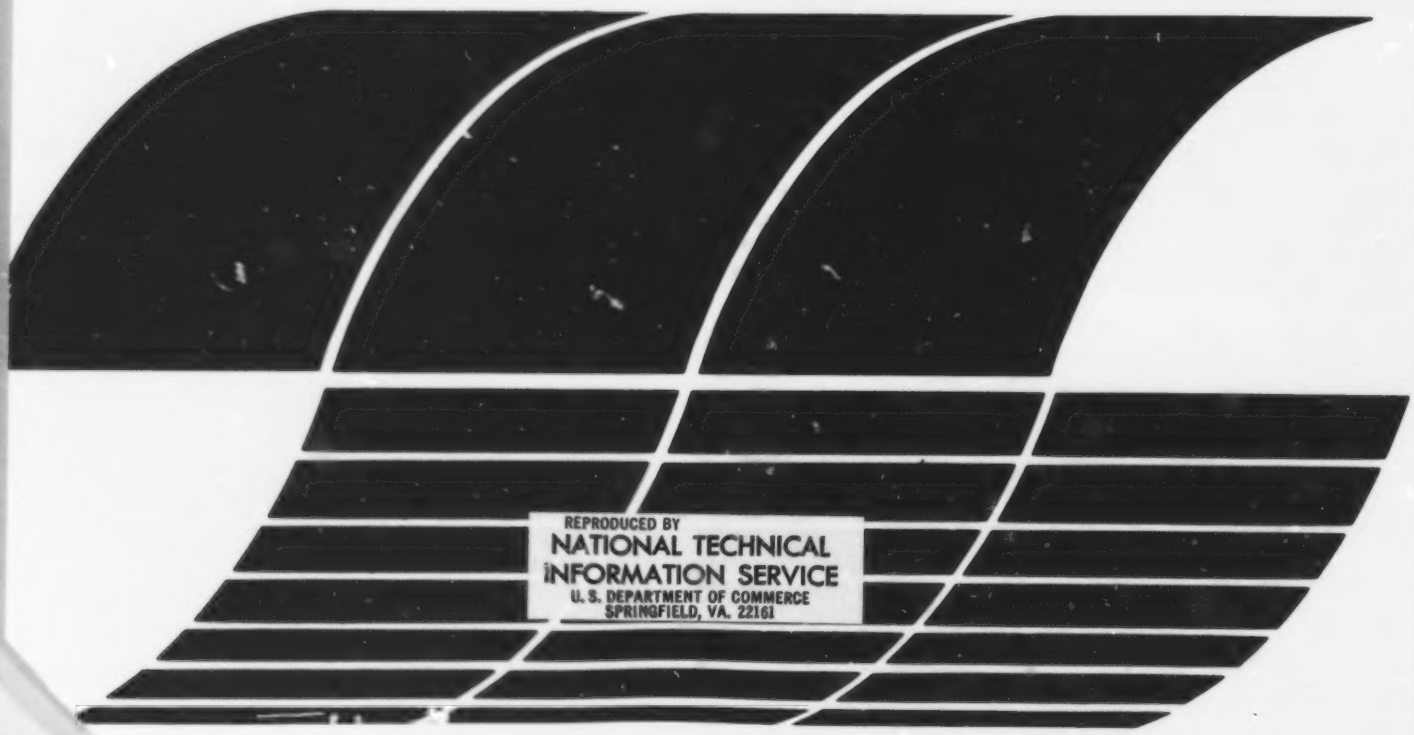

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PREFACE

The need to improve methods for evaluating the flux of airborne pollutants to vegetation and other natural surfaces is well appreciated. On the one hand, this flux represents one of the major sinks of atmospheric pollutants; on the other, it constitutes the dose to vegetation which is of importance in ecological studies of pollutant effects. Early investigations of dry deposition indicated that the rates of uptake at natural surfaces are often controlled by properties that are rather difficult to formulate, such as surface morphology and stomatal resistance. Moreover, the behavior of gases at interfaces depends to some extent on their solubility in water and reactivity with surface materials; quite different considerations enter into particle uptake.

Many wind-tunnel investigations served to identify most of the factors controlling the retention of pollutants by surfaces. But it is clear that the generality of such small-scale investigations is severely limited. In recognition of the need to extend investigations to field conditions, micrometeorological methods have been adapted to the case of pollutant fluxes. Most studies of this kind have relied on measurements of concentration gradients in the air, interpretation of which places great demands on the quality of the site selection, data-collection techniques, and analytical methods. To avoid some of the areas of uncertainty encountered in applying the gradient method, other micrometeorological methods have been employed. The most successful of these has recently proven to be the eddy-correlation techniques, which are described in this report and are compared with a number of competing methods.

ABSTRACT

Dry deposition on natural surfaces is one of the major removal pathways for air pollutants. In order to develop mathematical descriptions for the numerical simulation of the transport, removal, and ecological impact of pollutant gases and aerosols, the dependence of dry deposition rates on physical, chemical, and biological parameters must be understood. Such relationships can be studied by using several experimental methods to determine the vertical fluxes of pollutants over natural surfaces. The possible experimental methods include aerodynamic, modified Bowen ratio, eddy correlation, variance, and eddy accumulation. The relative advantages and disadvantages of these methods are discussed, with consideration being given to the sensor response time and accuracy. The roles of atmospheric stability and the zero plane, site and instrumental requirements, and averaging time are discussed for flux measurements.

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LIST OF ABBREVIATIONS AND SYMBOLS

a	Proportionality factor between pumping rate and vertical wind speed
C	Pollutant concentration
c_p	Specific heat of air at constant pressure
d	Zero-plane displacement height for momentum
E	Evaporation rate
F	Pollutant flux
H	Sensible heat flux
k	von Karman constant
L	Obukhov length scale; otherwise the latent heat of vaporization of water
m	Mass of pollutant accumulated
n	Frequency
p	Pumping rate
q	Specific humidity
r_{wC}	Correlation coefficient between w and C
Ri	Gradient Richardson number
T	Temperature; otherwise total sampling time
t_c	Exponential time response of pollutant sensor
t_w	Exponential time response of vertical wind sensor
u	Wind speed
u_*	Friction velocity
v	Volume
v_d	Deposition velocity
w	Vertical velocity
z	Height
Δt_c	Delay time associated with pollutant sensor
θ	Potential temperature
ρ	Air density
σ_C	Standard deviation of concentration fluctuations

σ_w Standard deviation of vertical velocity fluctuations

ϕ_H Dimensionless gradient of temperature

ϕ_M Dimensionless gradient of wind speed

ϕ_C Dimensionless gradient of pollutant concentration

SECTION 1

INTRODUCTION

Dry deposition of air pollutants at the surface is recognized to constitute a major sink, on the average of importance roughly equal to removal by precipitation scavenging. Clearly, it is worthwhile to investigate the physical, chemical, and biological processes that affect dry deposition, in order to develop descriptions adequate for realistic numerical simulations of the transport and fate of pollutants. As a convenience for use in simplified numerical models, the concept of a deposition velocity v_d has gained popular acceptance. This quantity is defined in terms of the associated surface flux F and concentration C_z at height z above the surface in question by

$$v_d \equiv F/C_z. \tag{1}$$

This definition indicates that v_d depends on all of the factors that affect flux-gradient relationships in the atmosphere (e.g. height, wind speed, surface roughness, and atmospheric stability), as well as on a number of surface biological, chemical, and physical properties.

In considering the influence of different processes on the net flux of pollutants to a natural surface exposed in open air, there is considerable advantage in considering the electrical analogy sometimes used in agrometeorology. In essence, this analogy apportions the total resistance to transfer according to various contributing resistances, usually assumed to be in series. The aerodynamic resistance to transfer during neutral and stable conditions, for example, is readily shown to be

$$r_a = u_z/u_*^2, \tag{2}$$

where u_z is the wind speed at height z and u_* is the friction velocity. A slight modification is needed for unstable conditions. The friction velocity is a measure of the wind drag on the surface, and is defined in terms of the surface momentum flux τ by

$$u_*^2 \equiv \tau/\rho, \tag{3}$$

where ρ is air density.

Another resistance which enters into consideration is the surface resistance r_s, which is a measure of the difference in behavior at the surface between momentum and the quantity in question. Since this and other contributing resistances have recently been considered for some trace gases (1), they will not be discussed fully here. For the present, it is sufficient to recognize that the ability to reduce the overall problem into a number of easily visualized component resistances forms the theoretical basis for the experimental program that will be introduced in this preliminary report.

Most surface deposition studies have utilized gradient methods to evaluate the flux. However, better methods can be applied, provided suitable sensors are available; gradient data are sometimes difficult to interpret. Tantamount among problems concerning interpretation are (a) difficulties in evaluating the vertical diffusivity and (b) the need to assume some level for the appropriate zero-plane displacement height. It is not valid that all meteorological and air chemical quantities share the same zero plane displacement.

The present experimental plan calls for the application of modern micrometeorological methods of vertical flux measurement to the problem of dry deposition, in order to investigate the processes controlling the uptake of pollutants to natural surfaces in natural surroundings. To this end, a systematic survey of a variety of surfaces has commenced.

SECTION 2

CONCLUSIONS

For adequate evaluation of deposition velocities by use of flux gradient methods, it is necessary to use methods capable of resolving, with high accuracy, vertical concentration differences amounting typically to about 9% in the case of v_d = 1 cm s^{-1}, and about 0.9% if v_d = 0.1 cm s^{-1}. For use in conjunction with eddy-correlation apparatus, similar percentage values apply to the sensitivity of the sensor. In the former case, it is required that the instruments be essentially drift free over considerable periods. In the latter case, a total response time of less than one second is usually required, but some drift can be tolerated. If eddy-accumulation methods were developed and applied, the response of the air pollutant sensor could be much slower. For all of these methods, sampling periods of 15-60 min are usually desired; site and sampling requirements for deriving accurate deposition velocities for use in the present studies are rather demanding and are not met by most air-quality monitoring stations. To date, standard eddy-correlation methods have proven successful for the cases of small particles, total sulfur, and ozone, but analyses have not yet been completed.

SECTION 3

RECOMMENDATIONS

The success of the preliminary studies described here underlines the need for new pollutant sensors to be developed. The criteria for an adequate sensor for eddy-correlation studies can be specified as listed below.

(a) The exponential time response evaluated for a 10% step function change and the delay time (i.e. the lag time between an imposed step function change in concentration and the appearance of a corresponding signal at the output) should be small. The values of the time response and the delay time should be separately known, and their sum should be less than 1 s for daytime work, 0.3 s for nighttime work.

(b) The sensor should be constructed such that obstruction to the natural air flow is minimal. In practice, the use of short sampling tubes and high flow rates provides a means for satisfying criterion (a) but long tubes may be needed to minimize interference by the sensing device itself.

(c) The sensor should be small. In particular, the largest dimension of the sensor ideally should not exceed more than about 10% of the height above the surface. For simple operations, this indicates a maximum dimension of no more than 40 cm.

Finally, differences already found over various surfaces indicate that testing should take place over a number of different kinds of surfaces typically found in regions of importance. Certainly, the common assumption that v_d is constant for a particular species of pollutant cannot be accepted without considerable caution.

SECTION 4

EXPERIMENTAL METHODS

Although most field studies of pollutant deposition have utilized some form of aerodynamic (gradient or profile) methods in order to evaluate vertical fluxes, this is but one of a number of techniques that are commonly used in micrometeorology. Here, some of these methods will be reviewed. A more thorough review of some of the methods can be found in Kanemasu et al. (2). The intent is to provide an indication of the relative advantages and disadvantages of alternatives, in order to assist in deciding on the best way to apply specific sensors with known performance characteristics.

AERODYNAMIC METHODS

In conditions of neutral atmospheric stability, the flux-profile relationship can be written in terms of local gradients as

$$F = ku_{*}z(\Delta C/\Delta z), \tag{4}$$

where z represents the geometric mean of the two heights employed. By division by the mean concentration C at height z, the deposition velocity can be found as

$$v_d = ku_{*} \cdot (\Delta C/C) \cdot (z/\Delta z). \tag{5}$$

In an experiment involving logarithmically spaced sensors with two-fold height intervals (e.g. at 1, 2, 4, and 8 m), the ratio $z/\Delta z$ is about 0.7. With a typical value of $u_{*} = 40$ cm s^{-1}, the gradient corresponding to a deposition velocity of 1 cm s^{-1} is found to amount to a 9% change in C. It is clear that the use of separate sensors to measure such a gradient will prove fruitful only if each sensor is capable of considerably better than 5% accuracy. Obviously, in the case of a much lower deposition velocity, much more stringent criteria might need to be satisfied.

In order to determine u_{*} in Eqs. (4) and (5), a gradient method requiring wind speed differences can be employed. For example, in neutral conditions the wind shear relationship

$$\Delta u = (u_{*}/k) \cdot (\Delta z/z) \tag{6}$$

can be used. This obviously leads to

$$v_d = k^2(\Delta u) \cdot (\Delta C/C) \cdot (z/\Delta z)^2. \tag{7}$$

Use of Eq. (7) in other than neutral conditions can be quite misleading, since strong stability effects on both Δu and ΔC are omitted. Wind speed differences (Δu) can usually be determined with sufficient accuracy with commercially-available cup anemometers if they are carefully calibrated in a wind tunnel by the users.

MODIFIED BOWEN RATIO METHODS

If an independent measure of one of the more common meteorological fluxes is available (i.e. the sensible or the latent heat flux), then problems arising from atmospheric stability can be eliminated by assuming equality of diffusivities. Using water vapor flux results in

$$F = E \cdot (\Delta C / \Delta q) \tag{8}$$

and using sensible heat flux results in

$$F = (H / \rho c_p) \cdot (\Delta C / \Delta T) . \tag{9}$$

Equation (9) is favored because temperature is usually measured more easily than moisture content. In the modified Bowen ratio methods, the demand for accuracy in the measurement of pollutant concentration is as great as with the use of profile methods; it is only the problems of atmospheric stability and of evaluation of the friction velocity that have been eliminated.

Suitable measurements of the latent and sensible heat fluxes are obtained easily during the daytime, either by inference from surface energy balance studies, by use of lysimeters, or by application of eddy-correlation methods. The use of eddy-correlation methods in determining pollutant fluxes is considered next.

EDDY-CORRELATION METHODS

These techniques evaluate the vertical fluxes as the sum of instantaneous products of the vertical wind component and the concentration of the quantity of interest. With primes to denote deviations from mean values and an overbar to show a time average, the eddy flux of a pollutant can be written as

$$F = \overline{w'C'} . \tag{10}$$

In the more-common cases of wind speed, temperature, and humidity, eddy-correlation methods are well-accepted for the measurement of the fluxes of momentum, sensible heat, and moisture, respectively. The natural frequencies of eddies that contribute to vertical fluxes near the surface of the earth can be scaled according to the height and wind speed, with the result that at high frequencies it is required that sensors respond fully to eddies of frequency approximately $n = u/z$ (e.g. ref. 3). Very slow fluctuations can be rejected in the analysis procedure and hence it is not necessary that the sensors be free from drift, provided appropriate filtering is applied to the signals. As a crude guide, we may assume that for use in wind speeds of 4 m s^{-1} at a height of 4 m above typical land surfaces in the daytime, it is

necessary for the sensor to respond fully to frequencies on the order of 1 Hz.

Many suitable sensors to measure the vertical wind component are available, ranging from simple propeller anemometers to relatively complicated sonic devices. This aspect of the problem is quite well understood. It remains to discuss the implications regarding the accuracy and sensitivity of the pollutant sensor. To do so, it is useful to examine the equation

$$F = r_{wC}\sigma_w\sigma_C, \tag{11}$$

which is essentially a definition of the correlation coefficient. It is known that $\sigma_w/u_* \simeq 1.3$ (q.v. ref. 4) and that the correlation coefficient r_{wC} typically takes a value of about 0.4 in the daytime. Manipulation of Eq. (11) then yields

$$v_d \simeq 0.5\, u_*(\sigma_C/C). \tag{12}$$

This implies that, in the hypothetical circumstances of the profile experiment considered earlier, a sensor for eddy-correlation use should be sufficiently sensitive to resolve fluctuations in concentration of about 5% of the average value (RMS).

VARIANCE METHODS

Equation (12) suggests an experimental method that might be suitable in some circumstances. Over open water surfaces, for example, it can be assumed on the basis of previous experiments that u_* is about 3-4% of the wind speed measured at a few meters height. Thus, with a sufficiently sensitive and noise-free instrument, deposition velocity can be estimated. However, since both r_{wC} and σ_w/u_* are dependent upon stability, Eq. (12) should be used only with considerable caution.

EDDY-ACCUMULATION METHODS

The difficulties inherent in developing fast-response chemical sensors for eddy correlation have led several groups to explore the feasibility of using the eddy-accumulation techniques of sampling. This usually calls for the use of a rapidly responding air switching and pumping system, driven by signals derived from a vertical velocity sensor in such a way that air is channeled to one container (filter or chamber) during updrafts and to another during downdrafts. The pumping rates p must be proportional by a known factor, a, to the magnitude of the vertical velocity over a specified time interval T. If mean concentrations C_1 and C_2 are measured after accumulation in chambers during updrafts and downdrafts, respectively, the vertical flux is

$$F = (C_1V_1 - C_2V_2)/aT, \tag{13}$$

where V_1 and V_2 are volumes in each chamber. These could be determined by direct measurement or by electronic means where $V_1 = \int_0^T p_1 dt$ and $V_2 = \int_0^T p_2 dt$ such that $p_1 = 0$ when $w < 0$ and $p_2 = 0$ when $w > 0$. Alternatively, if

samples of mass m_1 and m_2 are collected on a filter, the vertical flux is

$$F = (m_1 - m_2)/aT. \tag{14}$$

Some early work on this method addressed the case of water vapor, but in recent years most efforts have concentrated upon the extension of the method to carbon dioxide and air pollutants. Although a large amount of effort has been expended, only a few field results have been reported in the literature. Further, field experience supports the need for precise measurement of accumulated mass and for accurate alignment of the vertical velocity sensor. In the first case, it is easily shown that mass differences of the order of 1% need to be measured; this is about the same criterion as applies in the case of gradient measurement, so that systems which work well in gradient applications can also be applied in the present special case of the eddy-accumulation approach. In the second case, it is clear that any error in removing the mean value of the vertical wind component will appear as errors in the mass accumulated and result in a corresponding error in the derived deposition velocity. In an experiment performed in 4 m/s winds, for example, an error of 0.1 degree in the alignment of the vertical velocity sensor will result in an error of about 0.7 cm/s in the derived deposition velocity. Of course, if the output from the vertical wind sensor is filtered as it is in eddy-correlation methods employing analog circuitry, this error can be substantially reduced. This filtering is necessary for eddy-accumulation when it is not possible to orient an anemometer array to an accuracy better than about 1 degree in the vertical, such as in situations in which local surface nonuniformities cause flow patterns to change with wind direction. Filtering allows the vertical velocities to be measured relative to local streamlines, rather than to geographic vertical.

SECTION 5

ATMOSPHERIC STABILITY AND THE ZERO PLANE

THE ROLE OF ATMOSPHERIC STABILITY

A major aim of meteorological research in the past several decades has been to investigate the influence of stability on atmospheric turbulence near the surface. Stability affects the efficiency of the vertical turbulent transfer of pollutants, so that each of the aerodynamic methods described above will be affected as a result of heating or cooling at the surface (in addition to changing wind speeds imposed by synoptic-scale pressure gradients). In particular, every formulation which results from a manipulation of some flux-gradient relationship, such as Eqs. 5 and 7, should be modified if experiments are performed in non-neutral conditions.

The method generally accepted for introducing stability correction factors in profile relationships is the use of the dimensionless gradients

$$\phi_M = (kz/u_*)\cdot(\partial u/\partial z) \tag{15}$$

$$\phi_H = (\rho c_p k u_*/H)\cdot(\partial\theta/\partial z) \tag{16}$$

and, by analogy,

$$\phi_C = (ku_*/F)\cdot(\partial C/\partial z). \tag{17}$$

The results of recent experimental investigations of the forms of these relationships have been summarized by Dyer (5), who recommends the use of

$$\begin{aligned} \phi_M &\simeq (1 - 16\ z/L)^{-1/4}, && z/L < 0 \\ &\simeq 1 + 5\ z/L, && z/L > 0 \end{aligned} \tag{18}$$

and

$$\begin{aligned} \phi_H &\simeq (1 - 16\ z/L)^{-1/2}, && z/L < 0 \\ &\simeq 1 + 5\ z/L && z/L > 0 \end{aligned} \tag{19}$$

Here, stability is formulated in terms of the property z/L, where L is the Obukhov length scale of turbulence near the surface,

$$L \equiv -\rho c_p u_*\theta[(kg(H + LE/14)]^{-1}. \tag{20}$$

This definition of L includes a contribution by water vapor to buoyancy-included turbulence, incorporated by inclusion of the latent heat flux term, LE.

Algebraic manipulation of the basic equations shows that the stability parameter z/L is directly related to the well-known virtual gradient Richardson number via

$$Ri \simeq z/L, \; z/L < 0 \tag{21}$$

and

$$Ri \simeq (z/L)/(1 + 5\, z/L), \; z/L > 0 \tag{22}$$

Although it is well known that ϕ_H and ϕ_M differ significantly in unstable conditions ($z/L < 0$), there is no convincing evidence to suggest that ϕ_C differs markedly from ϕ_H. On the contrary, it is known that water vapor transfer can be characterized by much the same relations that apply in the case of sensible heat, and Sinclair et al. (6) have shown that the similarity extends to the case of carbon dioxide. In the absence of further definitive information, it seems appropriate to assume that $\phi_C = \phi_H$ is generally true for conditions ranging from moderately unstable to moderately stable.

The limits of our understanding of atmospheric turbulence are well exemplified by the relatively narrow range of stabilities over which Eqs. (16) and (17) can be applied with confidence. This range is limited to approximately $|z/L| < 2$.

THE ROLE OF THE ZERO PLANE

All discussion so far has been based on the assumption that the surface of interest is either bare or is covered by very shallow vegetation. However, with vegetation of significant height, z should not be taken to be the height above the soil surface. The zero-plane displacement height d can be visualized as a height where effectively all the momentum appears to be absorbed, according to the profile equations (momentum is actually absorbed thoughout the canopy). The height z is assumed to be zero at the distance d above the soil surface. As a rule of thumb, it is common to estimate the displacement height of the velocity profile as $d = 0.7h$, where h is canopy height. Analogously, a displacement height for heat can be found where an effective canopy surface temperature is defined and where all of the heat appears to be absorbed by surface materials. Such an analogy extends to pollutants, where the effective surface concentration is assumed to be that of air in contact with outer material surfaces at the zero-plane displacement height for that pollutant.

In practice, the assumption that the zero-plane displacement heights for momentum, heat, pollutants, etc., are the same can lead to considerable confusion and rather large errors in evaluation of profile and aerodynamic equations with the use of data collected above deep plant canopies. For example, consider the case of measurements taken above a dead corn canopy of mean maximum height 2.5 m above moist soil. The value of d would be near

1.75 m, but since the source of water vapor would be primarily near the surface, the displacement height for water vapor profiles might be considerably less than 1.75 m. In the early morning, when solar radiation mostly heats the top of the canopy, the displacement height for a temperature profile would probably be greater than d = 1.75 m; at high sun elevations, when solar radiation penetrates deep into the canopy, the displacement height might be less than d. Obviously, for the case of gaseous pollutants, the displacement height could be considerably less than 1.75 m if uptake takes place only at the soil surface and not with the rather inert, dried plant stalks and leaves. Further, particles might not be retained by dead plant materials as efficiently as by healthy vegetation, possibly causing changes in the displacement heights for a profile of sulfate particle concentrations.

Thus, considerable care should be exercised whenever assumptions regarding the heights of zero-plane displacements are important. In fact, there is some evidence in the scientific literature that profiles of trace gases, whose behavior has been assumed in the discussions above to be "similar" (i.e., that ϕ's are the same), are in fact not similar close to the tops of some canopies. We infer that the displacement heights might not be the same. One approach to overcoming this problem in application of profile equations is to use profile data sufficiently high above the canopy that the errors in the assumed displacement height produce a small relative error in values of z. For example, over a healthy corn canopy, the use of profile data above 4 m probably would produce only small errors in the estimation of fluxes. However, as discussed in the next section, fetch requirements often do not allow use of data obtained at such relatively large heights.

At this time, it seems appropriate to reiterate that the eddy-correlation method, when properly applied, produces flux estimates that are necessarily unaffected by all considerations of atmospheric stability and zero-plane displacement. It provides a proper evaluation of the vertical flux at a point in space; interpretation of these fluxes in terms of the underlying surface properties can then be addressed.

SECTION 6

FURTHER SITE AND INSTRUMENTAL REQUIREMENTS

Most micrometeorological techniques can be applied with confidence only when the site satisfies relatively demanding criteria concerning uniformity. Moreover, the horizontal extent of the site should be such that there is an upwind fetch of at least 100 times the greatest height of measurement (as measured from the appropriate zero plane). These matters are of considerable importance; if insufficient fetch is available, or if the site is inhomogeneous, local flux divergences might have an overwhelming effect. In cases in which uncertainties exist about the suitability of a specific site, surveys to test the extent of horizontal homogeneity are usually of great value. At times it is possible to apply relatively simple corrections for the effects of advection, for example, but considerable care should be exercised.

It is clear that each method of pollutant flux measurement imposes a specific set of criteria governing sensor performance and deployment. For example, the success of the gradient method depends on the ability to measure small differences with considerable accuracy; variance and covariance methods are inherently less sensitive to sensor drift or accuracy. However, both of the latter, turbulence measuring methods rely upon the use of rapidly responding sensors (typically 1 Hz or better), and this imposes a new set of difficulties which are not common to the gradient approach.

One effect of poor time response in sensors used for eddy correlation is the underestimation of fluxes. Quite often a sensor for measuring pollutant concentration consists of a reaction chamber through which air is forced after passing through a rather long sample tube. The exponential time response t_c evaluated for a small step change in concentration in the air entering the chamber is frequently an adequate description of response characteristics within the reaction chamber, largely as a result of the process of purging the chamber with air of changing pollutant concentrations. Both the response time and the delay (transit) time Δt_c, the time needed for a change in concentration at the sample inlet, located near the vertical wind sensor, to travel to the reaction chamber, should be minimized. When t_c and Δt_c are known, as well as sample height z, wind speed u_z, and stability conditions (z/L), the fluxes can be adjusted to values that would have been obtained if the sensors had ideal response. Corrections up to 30 per cent are not unusual, but confidence in larger percentages depends a great deal on knowing the response and delay times very accurately. Further, the spectral properties of the fluxes as a function of stability are not known with enough precision for large correction factors to be calculated accurately.

For example, consider the case of a propeller anemometer and a particle sensor used in neutral conditions above a moderately rough surface. We assume that the propeller has a response length of 2 m, so that the time response is 2/u, with no delay time. The various response and delay times assumed for the particle sensor are shown, with the results, in Table 1. The correction factors were numerically computed on the basis of techniques described by Hicks (7) and with cospectra presented by Kaimal et al. (8). Table 1 shows that an increase in Δt_c has about the same effect as an increase in t_c. As a rough guide, we apparently can assume that the percentage change in the correction factor is proportional to $u(t_c + \Delta t_c)/z$,

TABLE 1. EXAMPLES OF UNDERESTIMATES OF POLLUTANT FLUXES DUE TO POOR TIME RESPONSE OF SENSORS.

z	u	t_w	t_c	Δt_c	flux correction factor
m	m s^{-1}	s	s	s	
2	3	0.67	0.3	0.3	1.58
2	3	0.67	0.3	0.6	1.73
2	3	0.67	0.6	0.3	1.74
2	3	0.67	0.6	0.6	1.94
2	6	0.33	0.3	0.3	1.95
2	6	0.33	0.3	0.6	2.60
2	6	0.33	0.6	0.3	2.39
2	6	0.33	0.6	0.6	3.21
4	3	0.67	0.3	0.3	1.31
4	3	0.67	0.3	0.6	1.39
4	3	0.67	0.6	0.3	1.40
4	3	0.67	0.6	0.6	1.50
4	6	0.33	0.3	0.3	1.50
4	6	0.33	0.3	0.6	1.81
4	6	0.33	0.6	0.3	1.75
4	6	0.33	0.6	0.6	2.11
8	3	0.67	0.3	0.3	1.16
8	3	0.67	0.3	0.6	1.19
8	3	0.67	0.6	0.3	1.21
8	3	0.67	0.6	0.6	1.26
8	6	0.33	0.3	0.3	1.26
8	6	0.33	0.3	0.6	1.42
8	6	0.33	0.6	0.3	1.40
8	6	0.33	0.6	0.6	1.58

which at a value of about 0.45 yields the maximum correction that can be accepted with confidence, 30 per cent. Use of a vertical wind sensor with faster response does not improve the uncorrected estimates in this case in which a substantial time delay is assumed in the other sensor, because of

the effects of phase shifts induced by the lagged signal. It is expected that in slightly unstable conditions, the percentage error given by the correction factors can be reduced by about 20%, and, in moderately unstable conditions by about 40%. In stable conditions, the correction factors can become extremely large.

SECTION 7

AVERAGING TIMES AND DIURNAL CYCLES

Surface fluxes of any atmospheric quantity are a dynamic property capable of strong changes between successive sampling periods, particularly between day and night. Two aspects of this behavior are of special interest here: a) the time interval over which experimental data should be averaged, and b) the sampling time used in monitoring concentrations for the derivation of surface fluxes by application of experimentally derived deposition velocities.

SAMPLING TIMES FOR FLUX MEASUREMENTS

It is necessary for the sampling time to be long in comparison with the periods of turbulent fluctuations which contribute to the flux itself. Micrometeorological studies indicate that on these grounds sampling times should be longer than five minutes, and preferably greater than fifteen. Since conditions are required to remain steady during the period of averaging, periods of less than one hour are usually desired, although during the relatively stationary meteorological conditions around noon, considerably longer averaging periods might be permissible. Obviously, averaging through the transition periods near dawn and dusk should be avoided, since interpretation of the data obtained would present a number of severe difficulties.

The above comments on averaging times apply to experimental studies in which it is desired to investigate details of the deposition mechanism. If the intent of a particular study is simply to document the surface flux, such as for daily averages, then these sampling criteria obviously can be relaxed.

SAMPLING TIMES FOR MEAN CONCENTRATION MEASUREMENTS

Because atmospheric conditions change greatly throughout each diurnal cycle, resulting in large variations in deposition velocities, it is exceedingly difficult to derive accurate surface fluxes from concentrations averaged over more than a few hours. As an example, the case of pollutants emitted from a low-level source can be considered. Strong vertical mixing during the day will result in decreased concentrations near the surface, but surface fluxes might still be high. At night, low-level concentrations might be very high, but exceedingly small vertical diffusivities might lead to very low deposition rates.

In order to deduce surface fluxes from low-level concentration measurements, data must be obtained over averaging periods that are about the same

as are used for eddy flux measurements. If only a rough estimate of daily deposition is needed, it might be appropriate to consider concentration measurements made over periods as long as six hours, arranged so that the transition periods at dusk and dawn are excluded. It is necessary that the meteorological conditions be well defined, and that the site in question satisfy the usual micrometeorological requirements for horizontal homogeneity. It should be emphasized that few air-quality measurements meet the relatively demanding micrometeorological criteria. Thus, compliance with the rather stringent sampling guidelines implied here might not be possible in any but very rare cases.

SECTION 8

RECENT EXPERIMENTAL APPLICATIONS

Since contaminant fluxes can be measured directly with little ambiguity by use of the eddy-correlation technique, efforts have been made to apply this technique to the measurement of dry deposition of pollutants. Eddy correlation measurements permit the elimination of added steps such as the correction for atmospheric stability and indirect estimation of the friction velocity. Two past field experiments have applied the eddy-correlation method, with apparent success, to the cases of small particles (9) and ozone (10). In both cases, the simple analog flux evaluation methods developed originally for the real time evaluation of the fluxes of momentum, moisture, and sensible heat were adapted by subsitution of appropriate concentration sensors for one of the meteorological sensors.

During a study of pollutant fluxes to a forest of loblolly pine, conducted during July 1977 in collaboration with workers from the U. S. EPA and Duke University, eddy fluxes of small particles, ozone and total atmospheric sulfur were measured. No great difficulties were encountered in determining the eddy fluxes. The case of the ozone flux can be used to illustrate some of the points made above. Let us assume an ozone deposition velocity of 0.5 cm s^{-1} and a wind speed of 2-3 m s^{-1}, as was found in some cases. Then the ozone concentration change with height in constant flux conditions will be about 3% between the logarithmically-spaced levels 1-2 m, 2-4 m, 4-8 m, etc. If it were desired to determine the deposition velocity to 10% accuracy, it would then be required to evaluate each concentration to better than 0.3%, which seems rather prohibitive. Furthermore, the tacit assumption that the level of ozone destruction is the same as the zero plane for momentum transfer from the air seems to be particularly unpalatable since the canopy was so tall. Data obtained during this experiment will be presented in a separate document, as soon as analysis is completed.

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